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JCS42 U.S. PTO

## PROVISIONAL APPLICATION COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION under 37 CFR 1.53 (b)(2).

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The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

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Respectfully submitted,

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REGISTRATION NO.  
(if appropriate)

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☐ Additional inventors are being named on separately numbered sheets attached hereto

## PROVISIONAL APPLICATION FILING ONLY

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Attorney's Docket No. AN05982P**PATENT****IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**In re application of: Benyahia Nasli-Bakir, et al.Serial No.: 0 /

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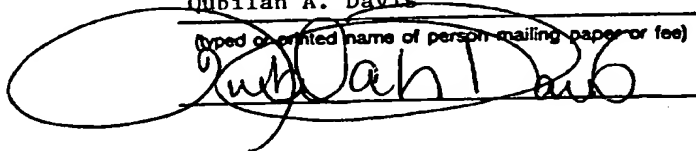
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## Method of application

According to the present invention a method of separate application of amino resin and hardener components, comprising a gluing system, onto a substrate, is provided. The present invention also provides a hardener composition comprising a volatile acid, which composition can be used in said method for improving the gluing result thereof, and a two-component gluing system comprising said hardener composition.

The resin component, preferably in the form of strands, is preferably applied first, whereafter the hardener component is applied. The method can, for example, be employed in the production of gluelam or laminated timber.

## Prior art

Separate application of the components in a gluing system on to a substrate, such as, for example, a piece of wood, is known in the art, and places certain restrictions on the characteristics of the components used. By keeping the different components, often only resin and hardener components, separated in the application device, many advantages is offered, as is known in the art. From the viewpoint of, inter alia, pot life, operation, and cleaning, separate application of the components of a gluing system is preferred over the application of a mixture of said components.

In EP 0 362 742 separate application of, inter alia, amino resin gluing systems to wooden parts is suggested, by means of curtain application of both the components, or of one component, and strand application of the other. However, there is no indication of a preferred order of application of said components. In the example given, only a phenol-resorcinol-formaldehyde resin gluing system was used.

WO 97/29161 is concerned with a two-component glue system for the production of laminated wood panels, and only in general

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terms mentions separate application of melamine glues. Neither a particular method nor a specific order of application of the components is suggested. The hardener component used contains 20-40 % by weight of an inorganic filler.

However, in practice, it has now been found that for separate application, when only a small amount of hardener is used, as is often the case, curtain application is difficult to realize with a good gluing result, since it is hard to secure the accomplishment of a continuous and uniform curtain formed of only a relatively small flow, possibly leading to an impaired gluing result and glue joints having reduced strength. Additionally, the curtain is easily affected by even small wind blows. Moreover, it has also been found that a high amount of filler in the hardener component, such as 20-40 % by weight, inhibits the achievement of an adequate blending of the applied components on the substrate.

Accordingly, the present invention provides a method of application of an amino resin gluing system according to claim 1, whereby the above-mentioned problems are overcome. Also, a hardener composition for amino resin gluing systems with a considerably lower content of filler is provided according to the present invention which suitably can be used in said method.

The amino resin used in the method of the invention can be any amino resin, such as, for example, urea-formaldehyde, melamine-urea-formaldehyde, melamine-formaldehyde, melamine-urea-phenol-formaldehyde resins, and furfuryl alcohol modified varieties thereof, the preferred resin being melamine-urea-formaldehyde.

According to the method, an amino resin, and a suitable hardener is applied separately to wooden substrates. In the method, the resin component is preferably applied first in the form of strands, whereafter the hardener is applied in the form of strands, or alternatively, is applied by means of spraying. By use of the preferred order of application according to the

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invention, any damages to the substrate, such as wood, due to the hardener composition is minimized.

As used here, the term "strand" also comprehends the meaning of the term "ribbon", also conventionally used in the art, and any other like term.

A hardener composition is also provided according to the present invention for use in combination with amino resin gluing systems, which composition advantageously can be used in the method of the present invention in order to make the gluing system faster-curing, and to obtain joints endowed with higher strength.

As an essential constituent of the hardener component in amino resin gluing systems, an acid is included. When aggressive acids, such as mineral acids, are used, remaining acid in the glued product might cause detrimental effects to the substrate or joint, reducing the strength of the substrate and/or the joint, and/or discolouration of the substrate by the acid, which is highly undesirable.

Thus, the hardener composition according to the present invention comprises a volatile acid, which can readily be evaporized so that no detrimental amounts of acid will remain in the resulting glue joint, and which also allows for relatively large amounts of acid (and thus hardener) to be used in order to obtain a fast-curing system, if desired.

According to the method of the invention, the hardener component can also be applied first, followed by the resin component, while the preferred order of application being the hardener component first, followed by the resin component.

Besides the volatile acid, the present hardener can also comprise water, thickener, resorcinol, and, optionally, a relatively low amount of filler.

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The hardener composition to be used in the method according to the invention comprises:

- up to 50% by weight of a volatile acid, preferably 10-30 % by weight, and
- less than 20% by weight of a filler.

As used here, the term "volatile" is to be understood as meaning; having a low boiling point, and/or having a low vapour pressure at room temperature.

Suitable acids are, for instance, organic acids, such as formic acid, acetic acid, etc.

The amount of filler used, if any, can be determined as suitable under the specific circumstances from time to time. However, as shown in Example 1, below, the strength of the resulting joint appears to be reduced with higher amounts of filler.

#### Example 1

A beam was prepared from from lamellae having the following dimensions:

length = 280 mm  
thickness = 33 mm  
width = 145 mm

To the hardener used in this Example, comprising 17 % by weight of formic acid, which had been added to water, kaolin in the amounts listed below was added as a filler, wherein the amount of 30 % is not an example according to the present invention, and, thereafter the hardener was thickened by means of polyvinyl alcohol to a viscosity of, at 25 °C, 2 000 mPas.

The resin and hardener components were applied separately in the form of strands, in the preferred order, in a total amount of 400 g/m<sup>2</sup>. The ratio of harder to resin used was 30:100, and the resin used was melamine-urea-formaldehyde.

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The time and temperature used, and pressure applied, in the pressing was 20 °C, 20 h, and 8 bar, respectively.

After pressing, and one day for the joints to stabilize, the beam was delaminated according to EN-391-B 1 cycle. The following results were obtained:

<u>Joint</u>	<u>Content filler in hardener</u>	<u>% Delamination</u>
1	0 %	0.0
2	5 %	2.0
3	15 %	6.1
4 (comparative)	30 %	24

A high proportion of acid makes the gluing system faster-curing. By using volatile acid, which easily can be evaporized, a relatively larger amount of hardener in relation to resin can be used, such as a ratio of hardener to resin of about 1:4-1:1, leading to a higher-strength joint and a faster-curing system, as shown in Example 2, below.

#### Example 2

A beam was prepared from from lamellae having the following dimensions:

length = 280 mm

thickness = 33 mm

width = 145 mm

The hardener used in this Example consisted of 17 % by weight of formic acid, which was added to water, and thickened by means of polyvinyl alcohol to a viscosity, at 25 °C, of 2 000 mPas.

Application and pressing were performed in the same manner as in Example 1, using the same resin.

After pressing and one day for the stabilizing of the joints, the beam was delaminated according to EN-391-B 1 cycle. The following results were obtained:

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<u>Joint</u>	<u>Ratio resin/hardener</u>	<u>% Delamination</u>
1	100:15	11.5
2	100:20	12.0
3	100:30	0.0

In the method of the present invention, the resin and hardener components can be applied in any order by conventional means for application in the form of strands or by means of spraying, more suitably, the resin and hardener are applied in the form of strands, or, alternatively, the hardener component can be applied by means of spraying, wherein the hardener in either case preferably is applied following the application of the resin.

However, when the avoidance of the contact of the hardener component with the substrate is of primary concern, such as, for instance, for protection of the substrate from the resin component, the resin component should desirably be applied so that the strands will form a continuous layer on the substrate, on top of which the hardener subsequently is applied in the form of strands, or by means of spray application.

In the case of application in the form of strands of both components, it is preferred that the hardener is applied on top of the resin strands, whereby any undesired contact of the hardener with the substrate can be prevented, and admixing of the components is enhanced.

Accordingly, in one embodiment of the method of the present invention, the hardener is applied on top of the resin strands. In this embodiment it is conceivable even to use hardeners, or hardeners having constituent(s), from which the substrate desirably should be protected, without damaging the substrate or impairing the strength of the resulting glue joint.

A suitable device which can be used in the method according to the present invention for the application in the form of strands of both the components, and especially in the above-

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mentioned embodiment of said method, is a device comprising a unit of at least two hollow members, at least one member for each component, provided with a number of orifices, from which orifices in each member the respective component is applied to a substrate below said hollow members, wherein the hollow members are connected to each other, said members being positioned above the plane of application, wherein each of the holes in one of the members are aligned in the machine direction with the corresponding holes in the other member(s).

By the use of such a device, when the resin component is applied first, the application of the strands of the resin component can also easily be adjusted, so that said strands, when applied to the substrate, coalesce to form an essentially continuous layer, on to which the hardener can be applied in the form of strands, or by means of spraying.

The application is conveniently carried out with amounts of the components to be applied to the substrate in the range of about 200-500 g/m<sup>2</sup>.

A suitable feeding rate is up to about 300 m/min, more suitably 30-250 m/min, and, preferably 60-200 m/min. At feeding rates below 30 m/min difficulties in dosage are likely to be encountered due to discontinuous strands emanating from the orifices (also referred to as dripping).

In this manner the substrate is protected from direct contact with the hardener, which is often highly desirable. The direct contact of the acid in the hardener, applied on top of the resin component on a first piece of substrate, with another piece to be glued together with the first one, is limited to a certain extent by the fact that some of the acid will have diffused, or migrated, into the resin component at the point of time when the pieces are brought together and pressed. This method allows for relatively large quantities of volatile acid to be employed.

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According to the present invention a two-component amino resin gluing system is also provided, the hardener in which system is comprised of volatile acid, and less than 20% by weight of filler. Said two-component amino resin gluing system can suitably be used in the method according to the invention. The preferred resin component of the system according to the invention is selected from the group of melamine-urea-formaldehyde, urea-formaldehyde, or melamine-formaldehyde resins, melamine-urea-formaldehyde resin being most preferred.

As stated above, the opposite order of application, ie., the hardener first, is also possible, although not preferred. This order of application can, for example, be used where it is desirable to avoid or minimize any emission of vapour of the acid into the surrounding atmosphere, such as, in the working environment, especially if the acid used is not detrimental to the specific substrate. Said order of application could also be advantageous, eg., in the case where an expandable gluing system is used, in order to prevent the generated gas from escaping into the atmosphere.

The method of the present invention could also be used for the separate application of the resin and hardener components of an expandable amino resin gluing system wherein said hardener component is acidic, and the resin component comprises one or more gas generating substance capable of forming a gas when contacted with said hardener component, wherein the hardeners of the present invention also can be used. A suitable corresponding resin component could, for example, comprise such gas forming substance(s) in an amount in the interval of 0.1-10 % by weight, as calculated on the pure, active form of said substance(s).

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## Claims

1. Method of separate application of resin and hardener components of an amino resin gluing system on to a substrate, characterized in that the components are applied in the form of strands or by means of spraying, or any combination thereof, in optional order of application, wherein the first applied component optionally can coalesce to form an essentially continuous layer thereof, onto which the other component is applied.

2. Method according to claim 1, characterized in that the resin component is applied in the form of strands, whereafter the hardener component is applied by means of spraying.

3. Method according to claim 2, characterized in that the hardener component is applied in the form of strands.

4. Method according to claim 3, characterized in that the hardener component is applied in the form of strands on top of the resin strands.

5. Method according to any of the previous claims, characterized in that the gluing system is selected from the group of melamine-urea-formaldehyde, urea-formaldehyde, or melamine-formaldehyde resin gluing systems.

6. Hardener composition for use in the method of any of the previous claims, characterized in that it comprises:

- volatile acid, and
- less than 20% by weight of filler.

7. Hardener composition according to claim 6, characterized in that it comprises volatile acid in an amount up to 50 %, preferably 10-30 % by weight.

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8. Two-component amino resin gluing system comprising, as the hardener component, the hardener composition of claim 6 or 7.

9. Two-component amino resin gluing system according to claim 8, characterized in that it is selected from the group of melamine-urea-formaldehyde, urea-formaldehyde, or melamine-formaldehyde resin gluing systems.

10. Device for carrying out the method according to claim 3 or 4, comprising a unit of at least two hollow members, at least one member for each component, provided with a number of orifices, from which orifices in each member the respective component is applied to a substrate below said hollow members, wherein the hollow members are connected to each other, said members being positioned above the plane of application, wherein each of the holes in one of the members are aligned in the machine direction with the corresponding holes in the other member(s).

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## ABSTRACT

A method of separate application of amino resin and hardener components, comprising a gluing system, onto a substrate, is disclosed, and a hardener composition comprising a volatile acid to be used in said method for improving the gluing result thereof. Also disclosed are two-component amino resin gluing systems incorporating such hardener composition.

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